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PARTICLE-FILLED DENTAL COMPOSITE: SHRINKAGE INDUCED RESIDUAL STRESSES AND OVERALL MECHANICAL PROPERTIES

ONDŘEJ PREJZEK*, TOMÁŠ MAREŠ *

* Department of Mechanics, Biomechanics and Mechatronics
Faculty of Mechanical Engineering, Czech Technical University in Prague
Technická 4, 16607 Prague, Czech Republic
e-mail: ondrej.prejzek@fs.cvut.cz, http://mechanika.fs.cvut.cz/

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Abstract. This work is focused on numerical simulation of the shrinkage phenomenon and its influence on overall elastic modulus of the composite and it is supported by experimental verification of results. It has been motivated by experimentally observed discrepancy between elastic moduli in uniaxial compression and tension. Its main contribution is a hypothesis linking this discrepancy with the mentioned phenomenon of polymerization shrinkage. The residual stress field after shrinkage is computed and its effect on the tangent stiffness of certain material points is discussed. The finite element model is built in a parametric manner with varying volume fraction and distribution of particles. To simplify the irregular distribution of particles in the composite, the model employs a crystallographic analogy of lattices. The plasticity model used to simulate resins behaviour has critical influence on the results. It is hypothesized, that the residual stresses beyond the yield stress weaken the model response in the compressive direction. Presented hypothesis is proven using the simplest von Mises criterion and further explored using the more realistic Drucker-Prager criterion.

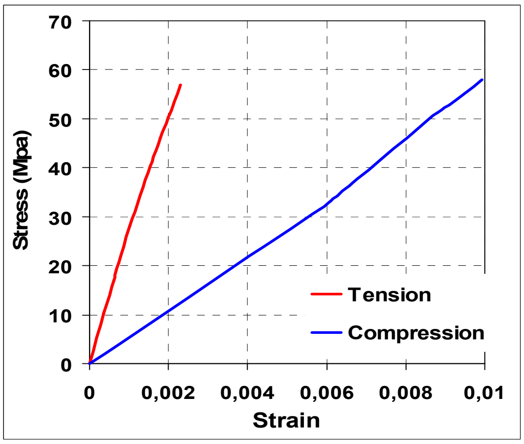
1 INTRODUCTION

Advanced dental materials belong to most common applications of particle reinforced composites. They consist from glass particles and polymer resin. Dental composites are used for their superior aesthetic properties and resistance to environment. However their mechanical properties, namely strength and resistance to fracture, still require to be enhanced. It is hypothesized, that certain mechanical properties are influenced by residual stresses developed due to polymerization shrinkage.

In order to predict fracture and obtain optimal shape of restoration to preclude its development, realistic modeling of the physical process of shrinkage and its effect on mechanical properties becomes critical.

Polymerization of the composite's resin causes its shrinkage, as the monomers are linked into chains [1, 2] which occupy smaller space. As the resin and entire composite material shrinks, internal residual stresses occur. Most of the literature, related to dental materials, investigates macroscopic stresses, developed in the composite, constrained by a tooth [3, 4]. There, the composite is considered to be homogenous material. Development of curing residual stresses on the interface between the filler and resin has been studied for ceramic/metal composites [5]. The influence of fiber arrangement in CFRP (carbon-fiber reinforced polymer) has been investigated [6], using the finite element method (FEM). Residual stresses in composite materials, occurring during polymerization, influence mechanical properties, such as strength [7], plastic behavior [8] or resistance to damage initiation and development [9].

During recent research on particle filled dental composites, discrepant values of Young‘s modulus have been observed, depending on applied measurement method. Tensile moduli are referred to reach values over 10 GPa, flexural moduli to be 6-8 GPa, and compressive moduli to be 3-5 GPa [10,11,12]. Elastic modulus of a composite, consisting of polymer resin (Ethoxyl-BPDMA, TEGDMA, Bis-GMA) and glass particles (SiO_2, BaO, B_2O_3, Al_2O_3) has been measured [13]. Results show significantly higher elastic modulus under uni-axial tension, than under uni-axial compression, although both particles and resin have equal modulus in both directions and yield strength of resin is higher under the compressive load (figure 1).



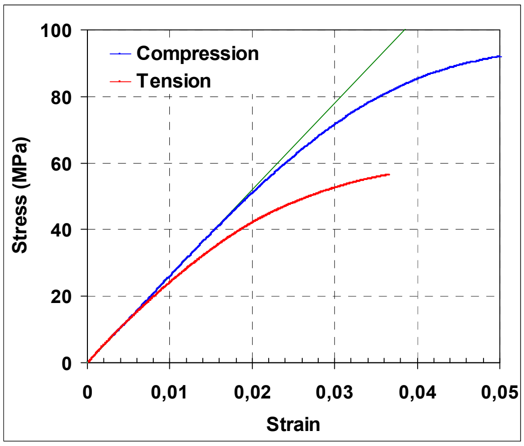


Figure 1: Stress-strain curves of composite (left) and resin (right)

This work tries to investigate a possible link between the polymerization, development of the residual stresses and the discrepancy between the elastic moduli under tensile and compressive direction.

2 NUMERICAL MODEL

The shrinkage process, development of residual stresses, and its influence on the overall response has been simulated using software ABAQUS. The amount of linear polymerization shrinkage of resin is referred to be up to 7% [2]. In the FE model, the shrinkage process has been mimicked by a thermal analogy. Regions occupied by resin have been prescribed coefficient of thermal expansion (CTE) of 7%, while CTE of particles is 0. The shrinkage strain, and consequently residual stress field, is introduced in the first step of analysis, by decrease of temperature by 1K. From second step of the analysis, external load in uni-axial compressive and tensile direction is applied, to simulate the test. Load is applied in 12 steps displacement of Δl/l by 0,05% in each, up to Δl/l=0,6%, what is behind the point, where the composite material breaks under tensile load in the experiments (figure 1).

In nature, the microstructure of the composite is irregular in size, shape and distribution of particles. In the model, particles are spherical, and its distribution is simulated by three crystallographic lattices, cubic, body-centered cubic (BCC) and face-centered cubic (FCC). For each lattice, simulation of five models is carried out, with mutual distance of particles varying from 1 to 20% of their diameter, while the volume fraction of particles reaches values from 30,3% (cubic array-20% gap) to 71,8% (FCC array – 1% gap). The microstructure is assumed to be periodical and is simulated by a Representative Volume Element (RVE) with defined periodical boundary conditions. To mesh the investigated microstructural unit, linear tetrahedral elements have been chosen for their low computational cost. The applicability of this particular element has been confirmed by an analysis of element-type.

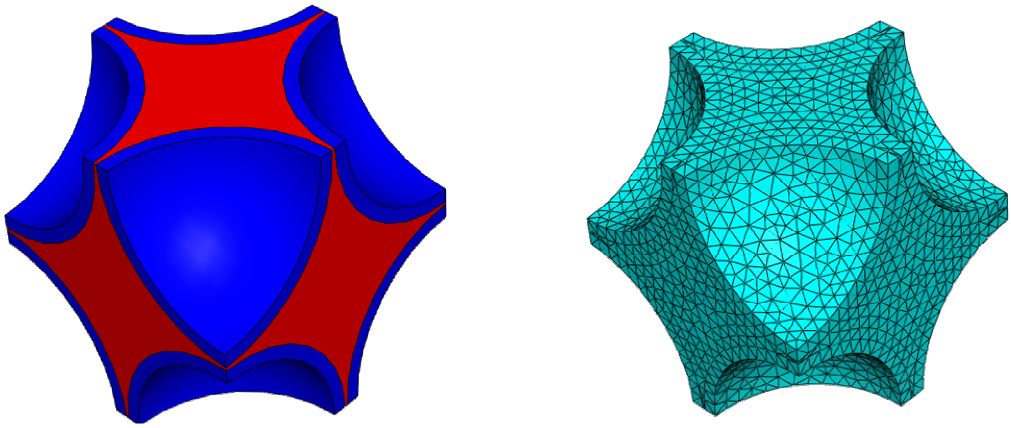


Figure 2: Cubic array – Unit cube and mesh

The modeled components of the composite material are polymer resin with an elastic modulus of 2,6 GPa and hollow glass particles with elastic modulus of 70 GPa. According to observations in [5], resin shows asymmetric yielding behavior with the initial yield stress of 20 MPa in tension and 50 MPa in compression, as can be seen in the Figure 1. Particles are considered to be perfectly linearly elastic. In first series of simulations, von Mises criterion, based on the lower (tensile) branch of the stress-strain curve has been applied. In the second one, the more realistic pressure-dependent Drucker-Prager criterion has been introduced.

3 RESULTS

3.1 Stress and strain distribution after curing – von Mises model

Distribution of stress and strain after polymerization curing has been studied. The distribution of residual stress (Figures 3,5,6) shows, that significant part of stress field is of a tensile nature, and only limited areas are compressed, in the sense of negative values of σ_{11}, σ_{22}, σ_{33}. If compared with the strain distribution (Figure 3), compressed are the regions, where the negative strain is higher than the value of shrinkage strain (7%). From Figure 4., development of plastic states can be observed. Compressed regions in narrow gaps between particles are highly plastic, while stretched regions remain elastic, or slightly plastic.

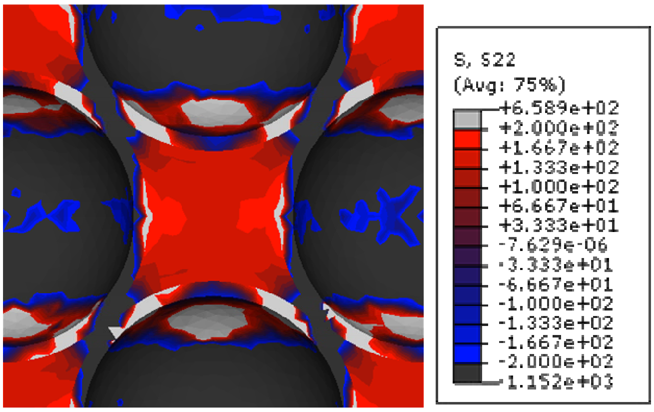


Figure 3: FCC Array-2% gap, von Mises material model: Distribution of σ_{22} after curing.

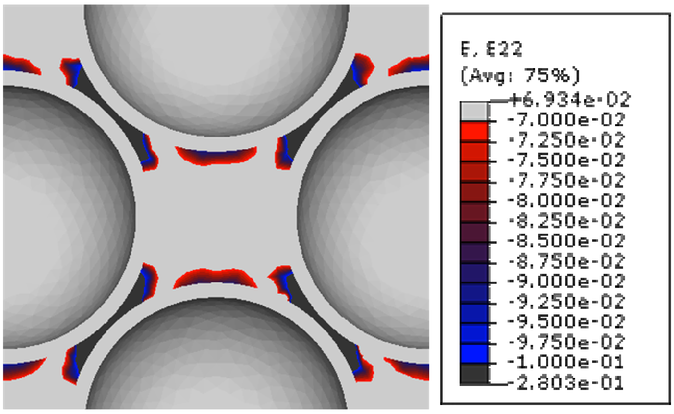


Figure 4: FCC Array-2% gap, von Mises material model: Areas with strain over the value of shrinkage-caused strain.

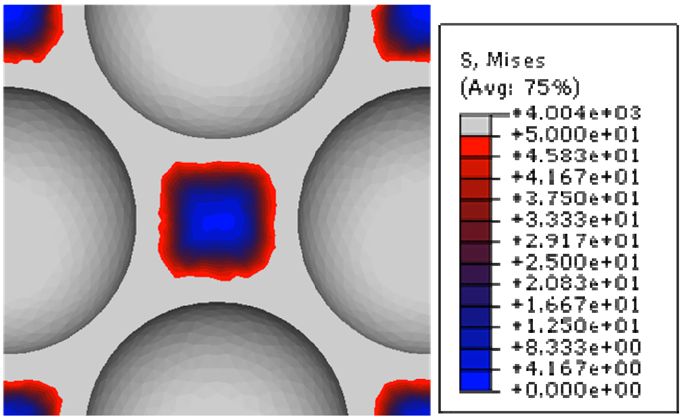


Figure 5: FCC Array-2% gap, von Mises material model: Elastic regions (σ_{eff} < 50 MPa) after curing.

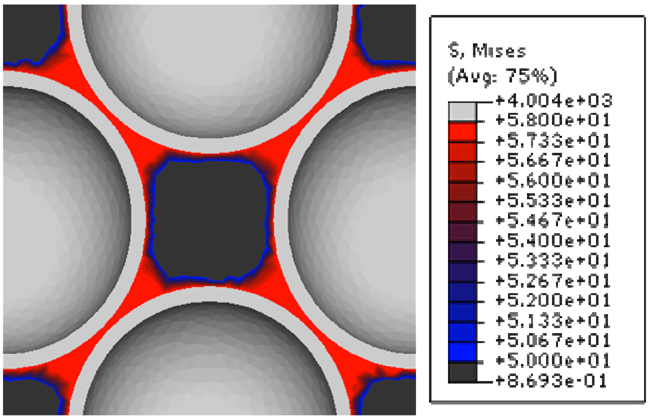
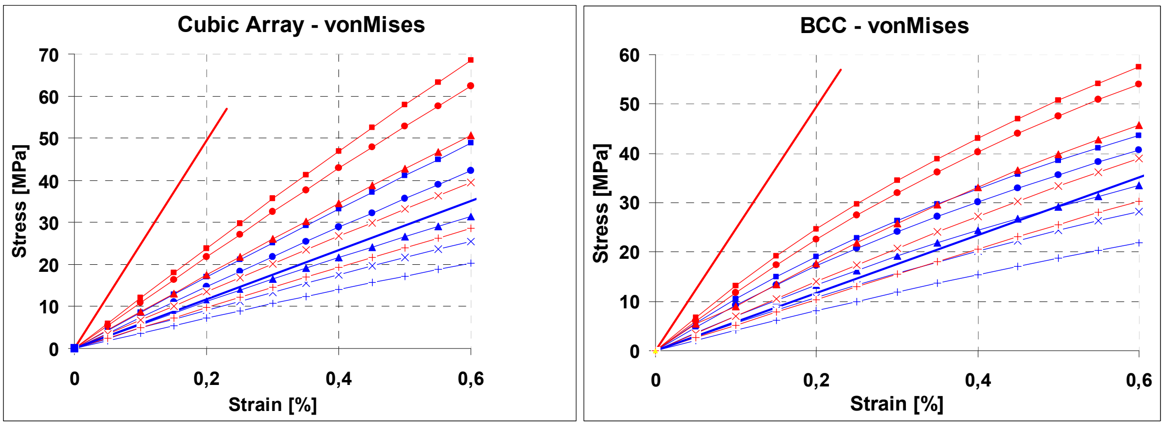


Figure 6: FCC Array-2% gap- Plastic regions (50MPa < σ_{eff} < 58 MPa) after curing.

3.2 Overall stress-strain response – von Mises model

The overall elastic modulus has been studied for each of the particle distribution arrays. Uni-axial tensile and compressive experiments have been simulated and resulting stress-strain curves (Figure 7.) have been compared with the results of experimental measurement. In the case of von Mises material model, for each of modeled arrays, the tensile stress-strain curve is higher than the compressive one, but the ratio of tensile (E_t) and compressive elastic modulus (E_c) does not reach the experimentally observed value. For each particular model, the E_t/E_c ratio is between 1,2 and 1,6 and the filler distribution does not tend to have influence on the ratio. Both tensile and compressive elastic moduli, by each of distribution arrays, tend to rise with the filer content. Except those models with very low filler content, all stress-strain curves are between the experimentally measured curves, and are closer to the curve of compressive experiment.



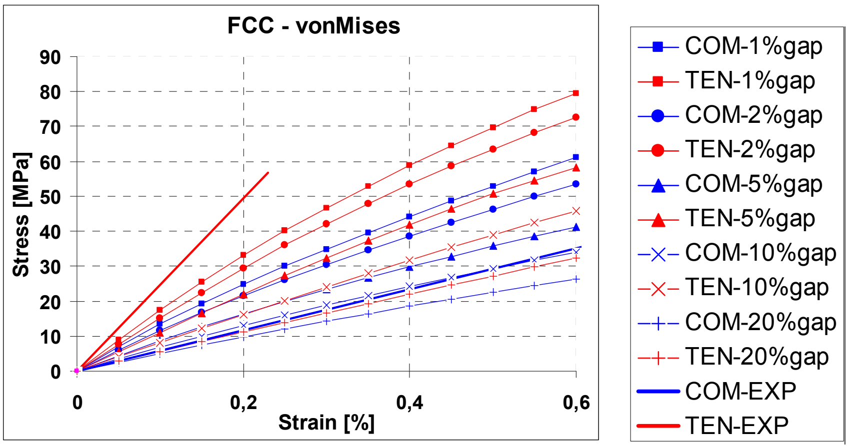


Figure 7: Stress-strain curves - Parametric simulation compared with experimental data, von Mises material model

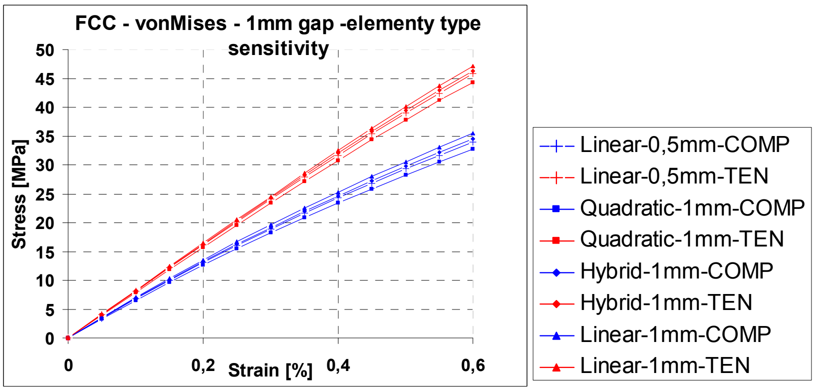


Figure 8: Stress-strain curves: Element type sensitivity (FCC-gap/particle diameter = 0,1)

Figure 8. shows the influence of chosen finite element on the results of stress-strain simulation. Linear and quadratic tetrahedral elements of various mesh density have been subjected to the analysis, as well as an element with hybrid formulation. The difference of elastic moduli in one direction does not exceed 10%, and affects both curves in the same manner, so the E_t/E_c does not significantly change. These results allow the use of linear tetrahedral elements, which have lowest demand for CPU time.

3.3 Stress and strain distribution after curing – Drucker-Prager model

Distribution of residual stress by the Drucker-Prager material model (Figure 7.) is of similar nature as observed by the von Mises model, while the plastic (in terms of the compressive stress-strain curve-Figure 1.) compressed regions are smaller (Figure 8.).

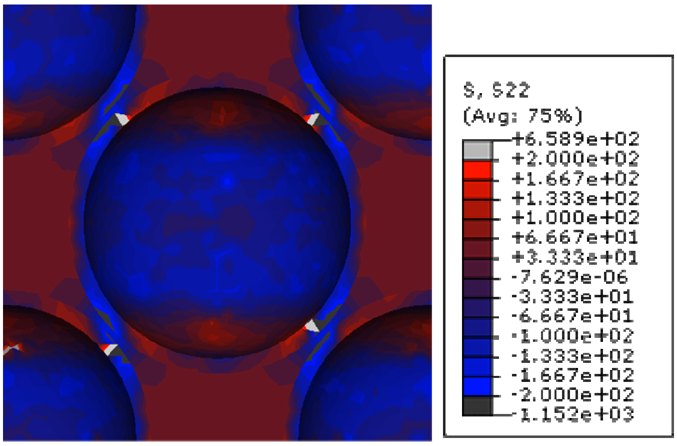


Figure 9: FCC Array-2% gapDrucker-Prager material model: Distribution of σ_{22} after curing.

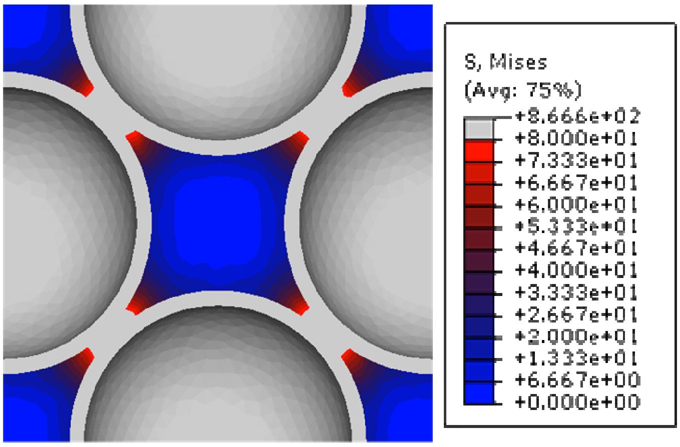


Figure 10: FCC Array-2% gap, von Mises material model: Elastic regions (σ_{eff} < 80 MPa) after curing.

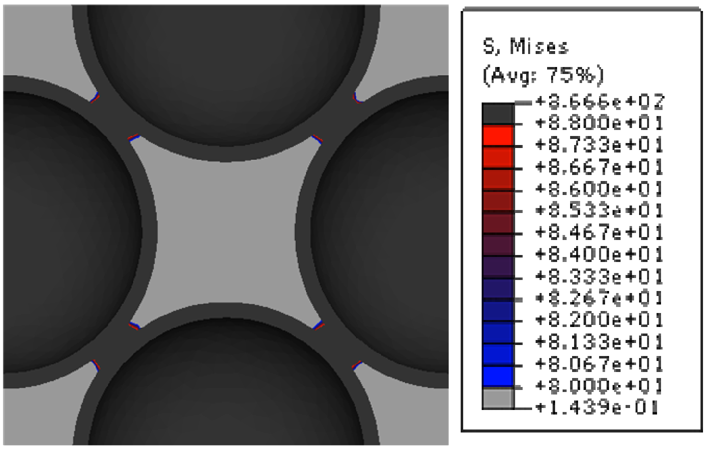


Figure 11: FCC Array-2% gap, von Mises material model: Plastic regions (80MPa < σ_{eff} < 88 MPa) after curing.

3.4 Overall stress-strain response – Drucker-Prager model

The stress-strain curves resulting from the simulation with the Drucker-Prager material (Figure 9.) do not capture the experimentally observed discrepancy of tensile and compressive moduli. At lower stress levels, the curves are close to equal, or the tensile curves are slightly higher. At higher stress levels, the tensile curves tend to be lower than the compressive ones. Resulting moduli are also higher than those of simulation with the von Mises material.

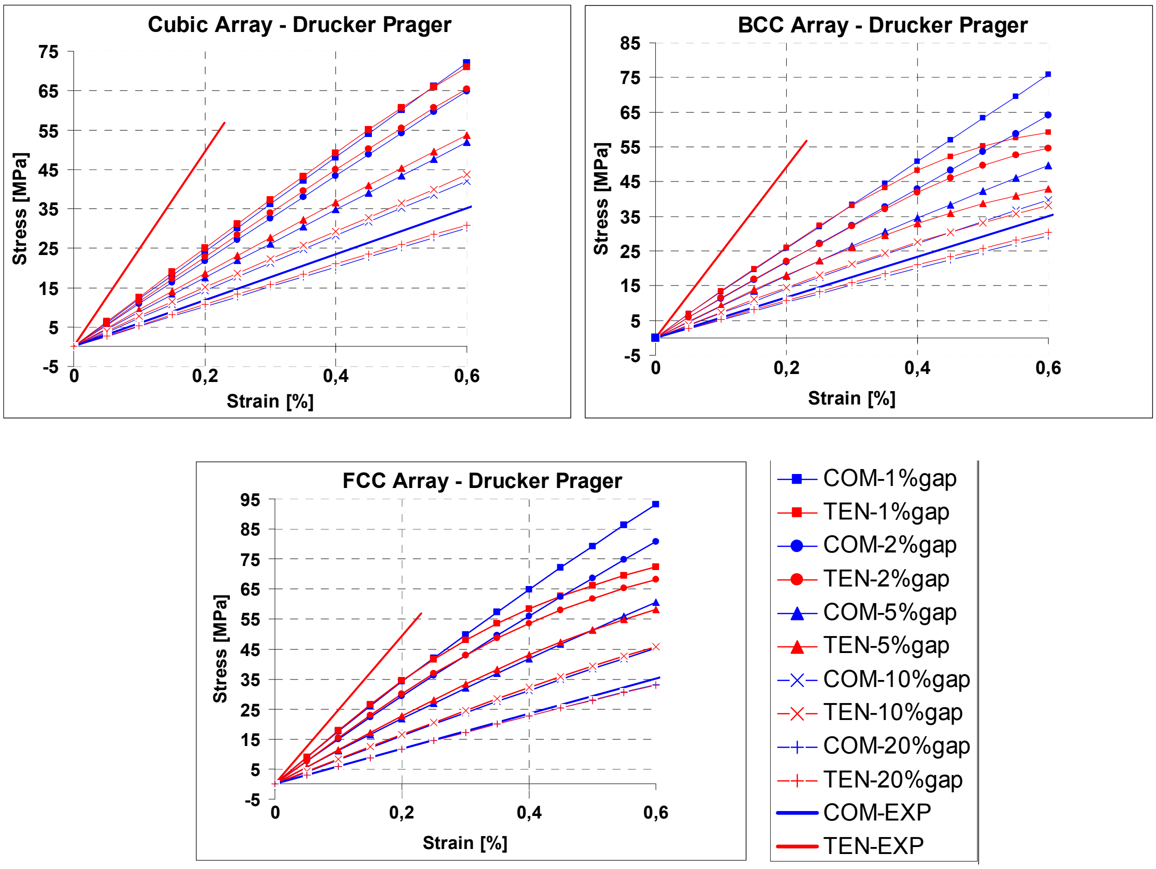


Figure 12: Stress-strain curves - Parametric simulation compared with experimental data, Drucker-Prager material model

4 DISCUSSION

All investigated models show similar distribution of stress in resin. In major part of resin, namely in large regions, the stress field is of tensile nature (Figure 3). Particles trying to move towards each other, due to shrinkage, are constrained by the shrinkage strain from the other side of particle, and the shrinking resin is stretched. In the regions of narrow gaps between particles, the particles moving towards each other compress the resin by more than the 7% of polymerization shrinkage. This causes the compressive stress in these regions. Similarity between stress distribution and plasticity development can be seen (Figure 3.,5. and 6.). Compressed regions are highly plastic, while stretched regions are elastic, or only slightly plastic. This fact mainly contributes to the discrepancy of tensile and compressive moduli. If the plastic, compressed regions are subjected to further compressive load, they respond with tangent modulus close to zero, while under tensile unloading their tangent modulus is equal to the initial elastic modulus. The tangent modulus of the tensile regions remains unchanged, elastic. Applying any mixing law, the resulting compressive modulus is lower than the tensile one. By the Drucker-Prager material model, the higher yield strength in compressive direction allows only small regions to become fully plastic, and smaller regions do not influence the model in that scale as by the von Mises model. This causes the minimal difference between E_t/E_c. The discrepancy between E_t and E_c can be better captured after implementation of the stress relaxation into the model. The tangent modulus of polymer resin after relaxation from plastic stress values is assumed to be close to zero under further loading and quasi-elastic under unloading. Then the compressed regions, with lower (relaxed) stress values must be larger to ensure equilibrium in the model, and they will contribute more to the weakening of the material in the compressive direction.

5 CONCLUSSION

Presented study shows the influence of residual stresses on mechanical properties of the investigated composite. Origin and nature of residual stress in particular regions of resin is explained as well as its effect on the overall elastic modulus. The highly plastic nature of the residual stresses requires an accurate model of plastic behavior. The simplest plasticity model, the von Mises criterion, shows how the residual stresses affect the composites behavior. The Drucker-Prager criterion is more suitable for modeling of polymers. In this particular case, it does not match the results and it needs to be enhanced by a stress relaxation model. In the future, stress relaxation properties of the resin will be measured and the data will be used to develop its visco-elastoplastic constitutive model.

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